
Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

LA-UR--83-2929

DE84 001719

TITLE: RELAXATIONS IN SPIN GLASSES: SIMILARITIES AND DIFFERENCES
FROM ORDINARY GLASSES

AUTHOR(S): K. L. Ngai, A. K. Rajagopal, and C. Y.

SUBMITTED TO: Proceedings of the 29th Conference on Magnetism and Magnetic
Materials, Nov. 8-11, 1983, Pittsburgh, PA.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.



By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

Los Alamos **MASTER**
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Relaxations in Spin Glasses: Similarities and
Differences from Ordinary Glasses

K. L. Ngai
Naval Research Laboratory, Washington, D.C. 20375

A. K. Rajagopal^{*}
Louisiana State University, Baton Rouge, LA 70803

C. Y. Huang[†]
Los Alamos National Laboratory, Los Alamos, NM 87545

ABSTRACT

Relaxation phenomena have become a major concern in the physics of spin glasses. There are certain resemblances of these relaxation properties to those of ordinary glasses. In this work, we compare the relaxation properties of spin glasses near the freezing temperature with those of glasses near the glass transition temperature. There are remarkable similarities between them and they both are in conformity with two coupled "universality" relations predicted by a recent model of relaxations in condensed matter.

^{*}Supported in part by ONR Contract N0014-82-K-0477.

[†]Supported by the U.S. Department of Energy

I. INTRODUCTION

There is a continued interest in the broad class of materials generally referred to as spin glasses which include the Cu:Mn, Pd:Mn metallic systems, some met-glasses, the $\text{Eu}_x\text{Sr}_{1-x}\text{S}$ insulating system and others. A common characteristic of spin glasses is the susceptibility cusp. This has led to the suggestion of a true phase transition to a novel phase. Another commonly observed feature of spin glasses is relaxation phenomenon which bears some degree of resemblance to relaxations in ordinary glasses and polymers. It has often been suggested the possibility that the spin glass transition is similar to ordinary glass transition. Relaxation phenomena are much better known and characterized in glasses than in spin glasses. Hence it would be beneficial to bring in what we know about relaxations in glasses, make intercomparisons with that spin glasses and deduce to what extent relaxations in glasses and in spin glasses are similar. This is one of the objective of this work. At this time, experimental investigations on relaxations in spin glasses is at the early stage. Although very beautiful data are emerging,¹⁻⁶ yet compared with glasses, the amount of knowledge is rather meager. Thus, to clarify ideas, we simulate for a model spin glass whose magnetic relaxation behavior near the freezing temperature T_f is ideally the same as relaxations in glass near T_g , the glass transition temperature. This can be done and is meaningful because by this time some general features of relaxation and their properties near T_g in glasses have been ascertained.⁷ Our considerations here are relevant independent of whether a genuine thermodynamic phase transition occurs at T_f or not. In either cases the frequency (time) dependence of relaxation needs to be considered and will be done here.

II. RELAXATIONS IN GLASSES

Relaxations in glasses remarkably conform to some "universal" patterns. As a function of time, relaxations are well described by the fractional exponential function

$$\phi(t) = \exp(-t/\tau_p)^{1-n}, \quad 0 \leq n < 1 \quad (1)$$

The statement holds for dielectric, optical, recombination, mechanical, volume, enthalpy, conductivity relaxations, etc., independent of whether the relaxation species is an electron, ion, dipole, molecular moiety, polymer segment or polymer chain. When formally interpreted as a "spectrum of relaxation times/frequencies" through the identity $\int g(\tau) \exp(-t/\tau) d(\log \tau) \equiv \exp(-t/\tau_p)^{1-n}$, larger n corresponds to a broader "spectrum." When the function in Eq. (1) or its time derivative is Fourier transformed, the complex dielectric susceptibility $\epsilon^*(\omega)$, electric modulus, $M^*(\omega)$, stress relaxation modulus $G^*(\omega)$, etc., can be readily obtained. Sample calculations of $\epsilon^*(\omega)$ can be found in Fig. 5 of Ref. 8. The peak frequency of ω_p , $\epsilon''(\omega)$, is approximately given by $1/\tau_p$. The primary relaxation responsible for glass transition conforms to Eq. (1) near, below and above T_g although n can be a function of T . Far above T_g , for nonpolymeric glass the fractional exponent n is zero or approximately zero and the effective relaxation time, τ_p , shifts with T in an Arrhenius way: $\tau_0 \exp(E_A/kT)$. Here τ_0 and E_A are respectively the attempt frequency and activation energy for an activated process and they both have physically meaningful values. As T approaches T_g , n increases and the "spectrum" broadens. At the same time τ_p as well as the mean $\langle \tau \rangle \equiv \int \tau g(\tau) d \ln \tau = [\tau_p / (1-n)] \Gamma(1/(1-n))$ depart from the Arrhenius behavior, increasing more rapidly in the manner as depicted in Fig. 1 for a glass. Here at several temperatures we give the mechanical relaxation data⁹ together with the fits to the prediction of Eq. (1). The variation of n as a function of T

is also plotted. These relaxation properties near T_g will be called here type A. They are generally observed for many glasses and polymers though not always. For example, there are glasses such as SiO_2 and GeO_2 that although they have non-zero n , yet it remains constant and τ_p shifts in an Arrhenius manner throughout the temperature range near T_g studied. We shall refer to these as type B glasses. Extensive review of data and analyses similar to that in Fig. 1 that establish empirically these regularities can be found in Ref. 7. In both types of glasses, τ_p and n are correlated and well satisfy another "universal" relation:

$$\tau_p = [(1-n)\omega_c^n \tau_0]^{1/(1-n)} \quad (2)$$

where ω_c is a characteristic upper cut-off frequency and τ_0 is the microscopic relaxation time of the relaxation species. Eq. (2) together with Eq. (1) are coupled predictions of a model.⁸ By coupled we mean that the same value of n occurs in both Eqs. (1) and (2). They have been tested extensively in many relaxation phenomena and found to be generally valid.⁷ For type A glasses, at high T , we have $n \rightarrow 0$ and it follows from Eq. (2) that $\tau_p \rightarrow \tau_0$. Hence, τ_0 can be identified with $\tau_\infty \exp(E_A/kT)$, at all temperatures. If n increases as T approaches T_g from above, then it can be seen from Eq. (2) that τ_p becomes non-Arrhenius even though τ_0 is Arrhenius. For a smooth monotonic increase of n , τ_p will have the qualitative features of the empirical WLF behavior¹¹ and the viscosity $\eta \equiv G_\infty \langle \tau \rangle$ may be well described by the empirical Vogel-Tamann-Fulcher (VTF) equation.¹¹ The solid line in Fig. 1 is the fit to the τ_p vs T data obtained by taking τ_∞ , E_A and $n(T)$ from data analyses and adjusting ω_c . Very near T_g , in some glasses including that shown in Fig. 1, η deviates from the VTF equation and reverts back to a steep Arrhenius behavior. In this regime, unfortunately often the relaxation time is so long that data of the exponent n for the relaxation process responsible for viscous flow are not

available. Below T_g , n depends on thermal history (cooling rate, annealing temperature and time, etc.)

III. RELAXATIONS IN SPIN GLASSES

That Eqs. (1) and (2) together with T variation (or lack of) of n to provide a good description of the relaxation of most, though not all, glasses, make it relatively simple to simulate for ideal spin glasses whose relaxation properties near T_f mirror those of glasses near T_g . Very recently Wenger⁴ have measured the ac magnetic susceptibility $\chi^* \equiv \chi' - i\chi''$ for an insulating spin glass $(\text{Ho}_2\text{O}_3)_{0.08}(\text{B}_2\text{O}_3)_{0.92}$ in the frequency range of $5\text{Hz} < f < 2 \times 10^3 \text{Hz}$ from 700 to 250 mK ($T_f \cong 420$ mK at 5.44 Hz). The data, limited though to less than 3 decades in f , do suggest that as the χ'' peak shifts to lower frequencies as T decreases its breadth remains relatively constant. The data can be fitted approximately by Eq. (1) with $n \cong 0.7$. No attempt is made to make an accurate fit because of insufficient data available at this time. Thus the holmium borate spin glass relaxation is of type B. We simulate its relaxation behavior by taking $n=0.7$ and T -independent, $\tau_\infty = 10^{-10}$ sec, $\nu_c = 10^{10}$ sec⁻¹, $E_A/k=2.1$ K and use Eqs. (1) and (2) to calculate χ^* . That is $\chi^*(\omega, T) = (C/T) \text{F.T.} \left[\left(\frac{d}{dt} \exp\left(-\left(\frac{t}{\tau_p}\right)^{1-n}\right) \right) \right]$ where F.T. stands for Fourier transform. In general, if there is some short range magnetic correlations among the spins above T_f , then the factor (C/f) should be replaced by the familiar $C/(T-\theta)$. From $\chi^*(\omega, T)$ we can obtain the conventional $\chi'(\omega_0, T)$ and $\chi''(\omega_0, T)$ for fixed ω_0 and variable T . The results are shown in Fig. 2. Bearing in mind that this is only a simulation with parameters arbitrarily chosen in the neighborhood of values suggested by experiment, it is remarkable that the features are in striking resemblance to Wenger's data. These include the relative sizes of χ' and χ'' , the shapes of the susceptibility cusps and χ'' peaks, the frequency dependence, the Arrhenius relation between ν and T_f with activation energy

8.0 K, and the sizable shift of the χ'' maximum to a lower temperature than $T_f(\nu)$ for each of the ν 's considered.

The Cu:Mn 5% spin glasses seem to conform to type A. Mezei and Murami⁶ have combined the neutron spin echo and ac susceptibility data to give a rough idea of how the relaxation changes with T. They found that at $\sim 4T_f$ the relaxation is exponential $e^{-\gamma t}$ with $\gamma=0.5$ meV. It continuously slows down as T is lowered. These data are consistent with a monotonic increase in n as $T \rightarrow T_f$ from above. Near T_f the decay is so slow that if Eq. (1) is obeyed, then very roughly, n is in the neighborhood of 0.9. Quantitative support for Eq. (1) comes from recent TRM measurements below T_f of Chamberlin⁵ which he found to obey accurately Eq. (1). The exponent n is about constant at low T up to $T/T_f < 0.8$ and thereafter increases monotonically to nearly 0.9 near T_f . In the insulating spin glass $\text{Eu}_{0.2}\text{Sr}_{0.8}\text{S}$, $\chi^*(\omega)$ data was obtained for the first time by Hüser et al.³ from 5.4 Hz to 2.8 kHz at fixed temperature in the range of 350 mK to 800 mK ($T_f=640$ mK at 10.9 Hz). The $\chi''(\omega)$ peak broadens markedly as T is lowered. The data being limited (e.g. peak of χ'' not clearly resolved, etc.) does not permit any accurate comparison with Eq. (1). Approximately, the data is consistent with Eq. (1) with n increasing gradually as T decreases from 800 to 700 mK, and then rapidly increasing from 700 to 600 mK. There seems to be a further gradual increase in n down to 350 mK to a value close to unity.

Both Cu:Mn and $\text{Eu}_x\text{Sr}_{1-x}\text{S}$ exhibit type A behavior. With Hüser et al.'s data in mind we simulate $\chi^*(\omega, T)$ through Eqs. (1) and (2) by assuming $n(T/T_f)$ to have T-variation as shown in Fig. 3, $\omega_c = 0^{10}$ radians/sec, $\tau_\infty = 10^{-10}$ sec, $E_A = 2T_f$ and $T_f = 800$ mK at 1.6×10^4 Hz. $\chi'(\omega)$ and $\chi''(\omega)$ are then calculated for various T. From these we can compute also $\chi'(T/T_f)$ and $\chi''(T/T_f)$ at various fixed ω . The results are displayed in Fig. 3 and they are remarkably similar

to the actual data. The $\chi''(T)/\chi'(T)$ ratio is smaller than that of holmium borate glass and the relation between ν and T_f is non-Arrhenius, resembling the VTF behavior.

IV. CONCLUSIONS

Analyses of various data taken in three different spin glasses enables us to conclude that relaxation properties near T_f of spin glasses resemble closely that of ordinary glasses near T_g . Holmium borate spin glass behaves like GeO_2 -type of glasses, while $\text{Eu}_x\text{Sr}_{1-x}\text{S}$ and Cu:Mn like $\text{Ca}_{0.4}\text{K}_{0.6}(\text{NO}_3)$ type glass. In spite of this difference, the relaxation properties of both types of spin glasses or glasses conform to two coupled "universal" rules predicted earlier.⁷ Simulations of χ' and χ'' by these rules, with parameters close to that suggested by experimental data, reproduces several features of the data rather well. We expect that other properties of relaxation in glass such as thermo-history dependence should be observable also in spin glasses.

REFERENCES

1. J.L. Tholence and R. Tournier, *J. de Phys.* 35C, 4-229 (1974).
2. J. Ferré, J. Rajchenbach and H. Maletta, *J. Appl. Phys.* 52, 1697 (1981).
3. D. Hüser, L.E. Wenger, A.J. van Duyneveldt, and J. Mydosh, *Phys. Rev.* B27, 3100 (1983).
4. L.E. Wenger, Heidelberg Colloquium in Spin Glasses, Preprint (1983).
5. R.V. Chamberlin, to be published.
6. F. Mezei and A.P. Murani, *J. Magn. Mat.* 14, 211 (1979).
7. K.L. Ngai in Non-Debye Relaxations in Condensed Matter, ed. T.V. Ramakrishnan, World Scientific Press, Singapore, 1983.
8. K.L. Ngai, *Comments Solid State Phys.* 9, 127 (1979); 9, 141 (1980).
9. R. Weiler, R. Bose and P.B. Macedo, *J. Chem. Phys.* 53, 1258 (1970).
10. C.A. Angell and L.M. Torell, *J. Chem. Phys.* 78, 937 (1983).
11. J.D. Ferry, *Viscoelastic Properties of Polymers*, 3rd ed. (Wiley, NY, 1980).

FIGURE CAPTIONS

FIG. 1. Plot of τ_p vs $1/T$ above T_g for $\text{Ca}_{0.4}\text{K}_{0.6}(\text{NO}_3)$. Δ , \times and \square are from experimental data (Refs. 9 and 10). \circ are predictions from Eq. (2). Insets are fits of Eq. (1) to mechanical relaxation data also from Ref. 9.

FIG. 2. Real and imaginary parts of χ^* obtained by calculation (see text for parameters) for the purpose of simulation of a type B spin glass: Holmium borate glass.

FIG. 3. Real and imaginary parts of χ^* obtained by calculation for the purpose of simulation of type A spin glass: $\text{Eu}_{0.2}\text{Sr}_{0.8}\text{S}$. The variation of n with T assumed is also displayed ($T_f=800$ mK).

Weller et al. (J.Chem.Phys., 53 1258(1970))





